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GASBUGGY

GAS QUALITY
INVESTIGATION PROGRAM
STATUS REPORT

**Lawrence
Radiation
Laboratory**
UNIVERSITY of CALIFORNIA
Livermore

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PNE-G-10

NUCLEAR EXPLOSIONS — PEACEFUL
APPLICATIONS (TID-4500)

GASBUGGY
GAS QUALITY INVESTIGATION PROGRAM
STATUS REPORT

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Lawrence Radiation Laboratory
Livermore, California

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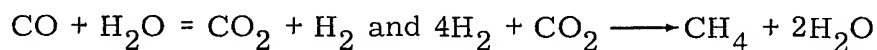
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GAS QUALITY INVESTIGATION PROGRAM STATUS REPORT FOR PROJECT GASBUGGY*

Abstract

Data relating to the chemical and radiochemical results from Project Gasbuggy chimney gas samples obtained prior to 210 days after the detonation are presented for significant non-radioactive components of the gas and for the tritium and Kr⁸⁵. A discussion of changes in composition occurring during the flaring of 5×10^7 ft³ gas in late spring 1968 is included. Some interpretation of the observed changes is advanced, but with the data now available, no definitive conclusions seem warranted. This is a status report of the continuing effort to define and understand the chemical and radiochemical aspects of project Gasbuggy.

Major constituents of the Gasbuggy gas during the period from 34 days to 200 days following the detonation were methane (increasing from 37% to 44%), ethane (increasing from 4% to 5%), propane (constant at ~1%), carbon dioxide (constant at ~36%), hydrogen (decreasing from 17% to 12%) and carbon monoxide (decreasing from 4% to less than 2%). Much more significant changes are observed during the first month. The major reactions used to explain these trends are:



Kr⁸⁵ concentration ($2.8 \mu\text{Ci}/\text{ft}^3$ NTP) remained essentially constant over the entire sampling period implying mixing with a constant volume (1.2×10^8 ft³ NTP) of noncondensable gas during this time.

Tritium was observed primarily as hydrogen at early times. A rapid decrease in HT came within the first month converting most of the HT to HTO, but producing some CH₃T and C₂H₅T. The predominant tritium-containing

*Work performed under the auspices of the U. S. Atomic Energy Commission.

species, except at very early time, is CH_3T at a concentration of 12 to $14 \mu\text{Ci}/\text{ft}^3$ NTP. Both CH_3T and $\text{C}_2\text{H}_5\text{T}$ concentrations increase slightly over the first 200 days, that of HT continues to decrease. The ratios $\text{CH}_3\text{T}/\text{CH}_4$ and $\text{C}_2\text{H}_5\text{T}/\text{C}_2\text{H}_6$ are essentially constant over the period from 30 to 200 days implying exchange equilibrium was attained rapidly. The ratio HT/H_2 continues to decrease over the same period implying a continuing influx of non-tritiated water into the chimney and a reasonably rapid exchange reaction between HT and H_2O .

Changes in concentrations of cavity gas components as a function of flow rate indicate that removal of 30% of the original chimney gas was accomplished by flaring $5 \times 10^7 \text{ ft}^3$ at a rate of $5 \times 10^6 \text{ ft}^3/\text{day}$. This result is encouraging, but the test was too short to provide verification of this process as a reasonable method of reducing contamination levels.

Introduction

Among the more important problems related to the application of nuclear explosives to stimulation of natural gas fields is that of radioactive contamination of the chimney gas. One of the primary objectives of Project Gasbuggy is "to determine the gas quality with regard to contamination by radioactivity, and to evaluate various techniques suggested for reducing this contamination."

A large quantity of data has been collected from analysis of the Gasbuggy chimney gas, and some systematic trends have been observed. Although some of the chemical and radiochemical analyses are not yet complete, that portion relating to the most important nuclides, T and Kr^{85} , and a body of extremely interesting chemical data are presented in this report. However, only a tentative assessment of these incomplete results is intended. More information must be assembled before detailed interpretations of the chemistry can be made.

Indeed, the lack of samples at early times when major changes were occurring may preclude unambiguous interpretation of the processes involved as far as this particular experiment is concerned.

Analytical Procedures

This section describes the process through which the data was obtained. Figure 1 is an overall view of the gas-analysis laboratory. The sample is introduced to the separation system via the inlet manifold shown on the far left, and condensed on the first large activated charcoal column. These separation systems are, in reality, very large scale gas chromatographs. Samples of Gasbuggy gas as large as $1/2 \text{ ft}^3$ can be easily separated. Figure 2 shows a closer view of the system. Helium is passed through the column as the temperature is progressively raised step-wise. The gases pass through the column in inverse order of their degree of adsorption. For a complete separation both charcoal (shown in the large cylindrical trap) and molecular sieve (shown in the W-shaped column trap) are employed at temperatures ranging from liquid nitrogen (-240°F) to $+600^\circ\text{F}$.



Fig. 1. General view of the gas analysis laboratory.

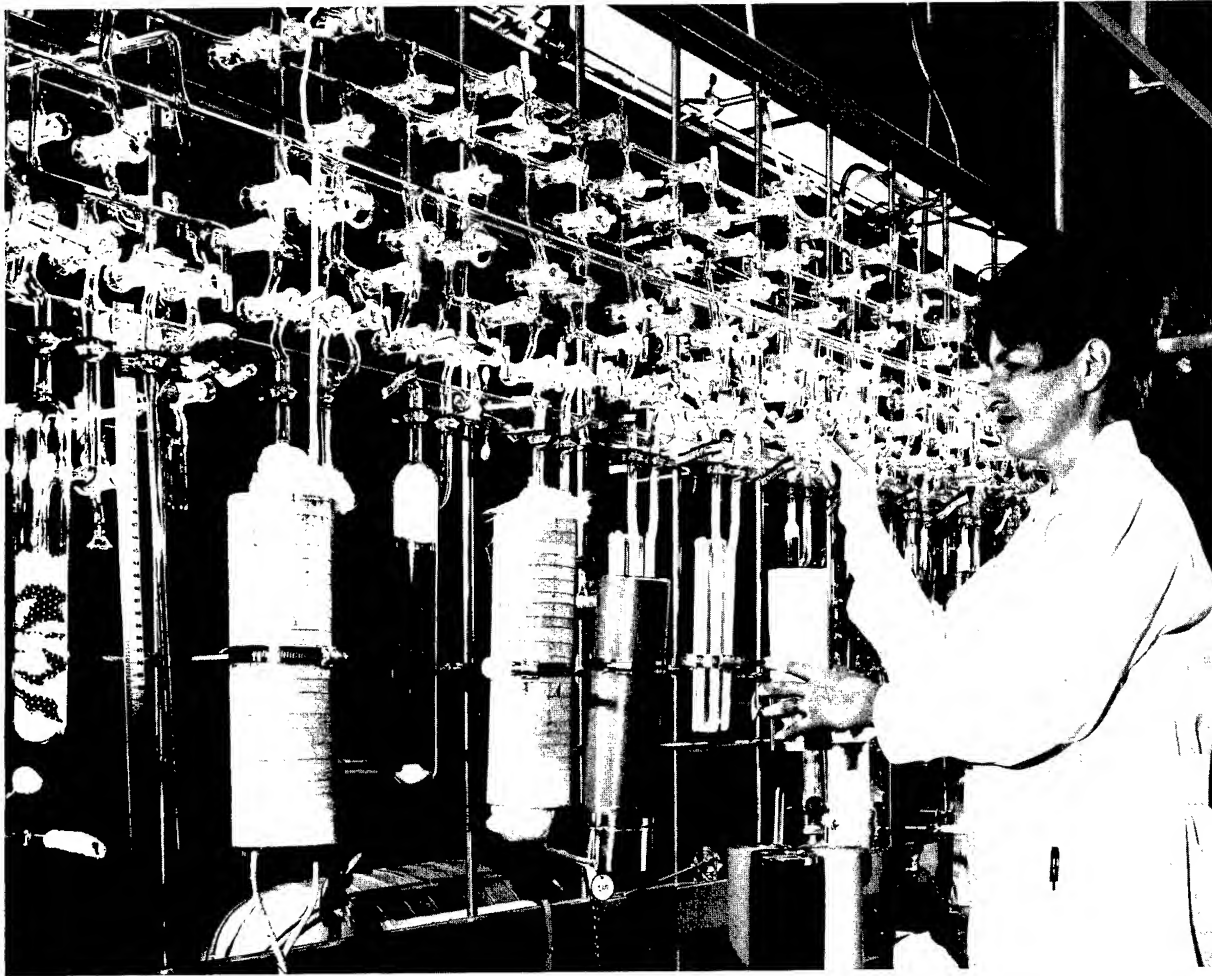


Fig. 2. Gas analysis separation system.

During the course of an elution, progress is monitored using a thermal conductivity detector and an ionization chamber. These are read out on the chart recorder shown in Fig. 3. Also shown is the sample recovery section of the separation system. The purified gas is recovered for transfer to a radiation counter.

Two types of counters are used. The "gas-cell" and thin window proportional counter is employed for Kr^{85} radio-assay. One of these systems is shown in Fig. 4. The krypton is contained in the small cells being loaded on the sample changer. Each cell, in turn, is rotated underneath the lead shield, where it is raised to counting position. Gases containing tritium are placed in internal proportional counting tubes, and become a part of the fill gas in the active volume of the tube. Figure 5 shows these tubes being inserted in counting position inside a shielded cave.

Uncertainties involved in counting are reduced by the standard practice of counting replicates. Results of duplicate counts are averaged to obtain the final result and an estimate of its reliability. The numbers to be presented here have individual standard deviations of less than 3%. Precision within a group of samples includes this uncertainty but is primarily determined by real variations in sample composition between samples. As will be seen, this variation is significant for hydrogen but much less so for the other gases of interest. In the data which follow error bars are used to indicate precision, standard deviation of the mean included. Where no such bars are shown they lie within the plotted point symbol. All data are related to cavity gas after air (based on O_2) was removed from the sample.

Chemical analysis by mass spectrometry was performed by R. Crawford of the LRL Analytical Chemistry Group. Analytical errors vary according to

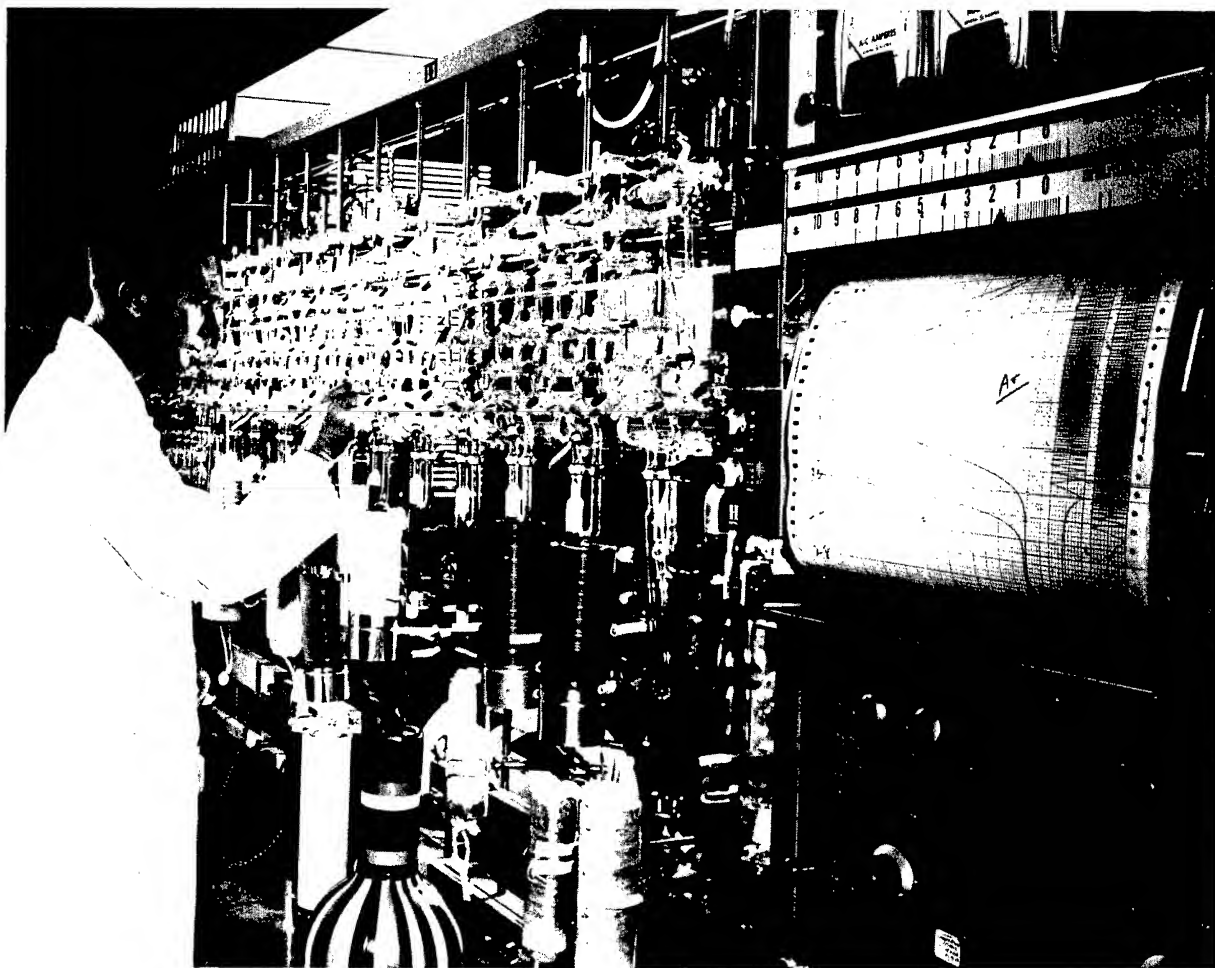


Fig. 3. Sample recovery section and chart recorder of the separation system.

the percent composition, but are generally less than 1 or 2 percent for the species of interest here. Small variations between samples within a group have been observed, and are the primary source of the errors attached to the data.

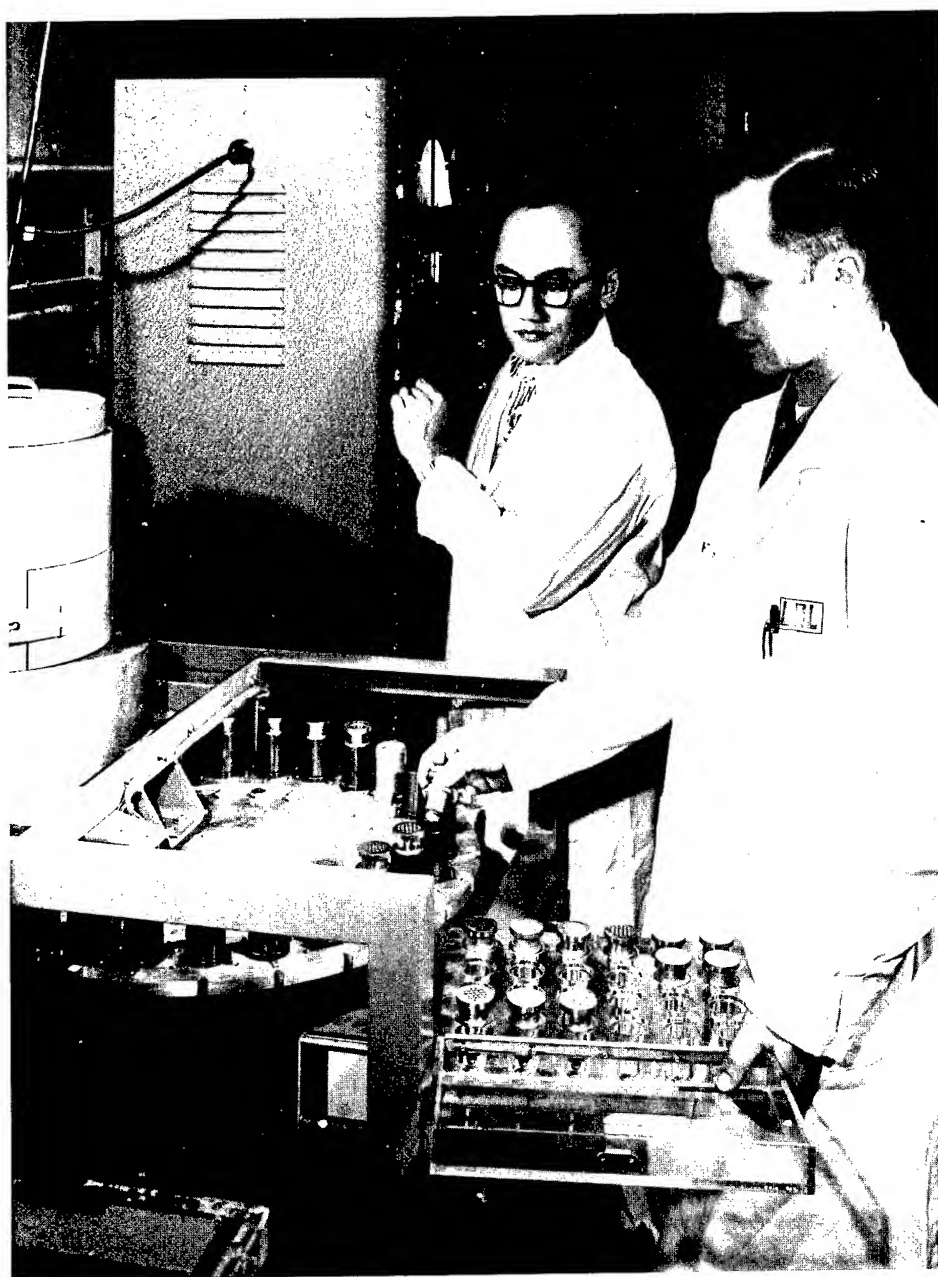


Fig. 4. Krypton radio-assay counter.

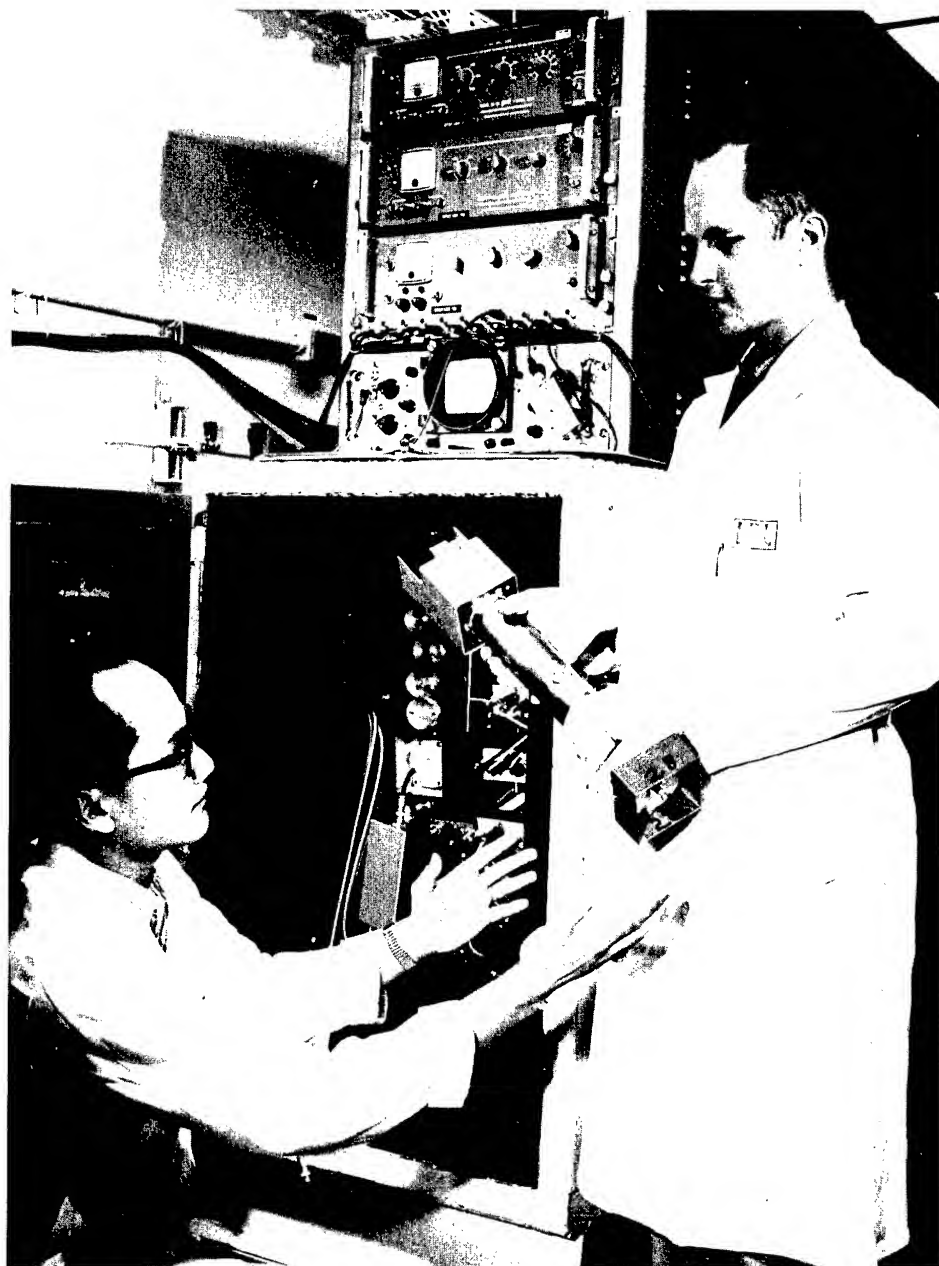


Fig. 5. Proportional counting tube for tritium being inserted in shielded cave.

Sampling

At the present time data are available from sixteen samples grouped in five sampling periods. Except for the production testing which occurred last June and July no significant variation was observed within a sampling period. Therefore samples within a period have been averaged. Only these averages are presented here. These averages are identified according to their sampling time mid-point as follows:

- 1 day: Four samples were obtained as a result of cable leakage to the sealed annulus of the emplacement hole. While these samples were gathered at about one day after the detonation, the actual time of their separation from the body of chimney gas is most certainly much shorter. They probably represent the chimney gas composition shortly after chimney collapse. Two were suitable for radiochemical analysis. However, these were 85% air. Therefore, the errors of the chemical analyses are magnified for the 15% of the sample deemed "cavity gas." In spite of this, the results do seem to provide useful information, and to fit well with the main body of information obtained from the other groups of samples.
- 34 day samples: Seven samples were taken between 32 and 36 days after detonation when communication with the chimney by GB-ER had been established. Five of these have been analyzed - two downhole samples and a surface sample taken before $0.3 \times 10^6 \text{ ft}^3$ gas was flared and one downhole and one surface sample after flaring. No significant differences in the results reported here were seen between surface and downhole samples, or between samples prior to and following the flaring. The air correction of sample composition to obtain cavity gas composition was a few percent for these samples.
- 79 day samples: Of the four samples taken, results from one surface and one downhole sample following flaring of $40 \times 10^3 \text{ ft}^3$ gas will be reported. As was the case at 34 days, no significant difference was observed. Downhole sampling has therefore been eliminated. The air correction was only 1% for these samples.

- 134 day samples: Results of the two surface samples taken after $4 \times 10^3 \text{ ft}^3$ were flared are included in this report. No air correction was required.
- 203 day samples: Data points at 203 days are taken from the first good sample obtained during the extended flow test. Approximately $5 \times 10^6 \text{ ft}^3$ of gas had been flared prior to sampling. Samples were taken at intervals of $5 \times 10^6 \text{ ft}^3$ or daily. Analytical results from six of these will be reported here. No air correction was required for these samples.

Gasbuggy Analytical Results

For convenience of presentation, the samples have been divided into two time-periods. The first group is composed of those samples taken during the shut-in period prior to flow testing. Samples obtained during the flow testing comprise the second group. Somewhat arbitrary curves have been drawn through the data points.

Figure 6 presents the observed changes in chemical composition of the cavity gas as a function of time. (These changes are also tabulated in Table I.) The effect of temperature equilibrium is clearly evident. Light gases predominate at early times, moving towards more complex gases as the chemical equilibrium shifts. Plotted across the lower portion of the graph is the total volume of gas with which the Kr^{85} is mixed. The constancy of the total gas volume within analytical uncertainty is remarkable. Evidently equilibration of the cavity with formation pressure occurred quite rapidly and has been maintained throughout the shut-in period.

The rapid increase in CO_2 and the corresponding decrease in CO suggests that the water-gas reaction:



is proceeding rapidly at early times.

The gradual decrease in H_2 concentration appears to be due to the reaction:

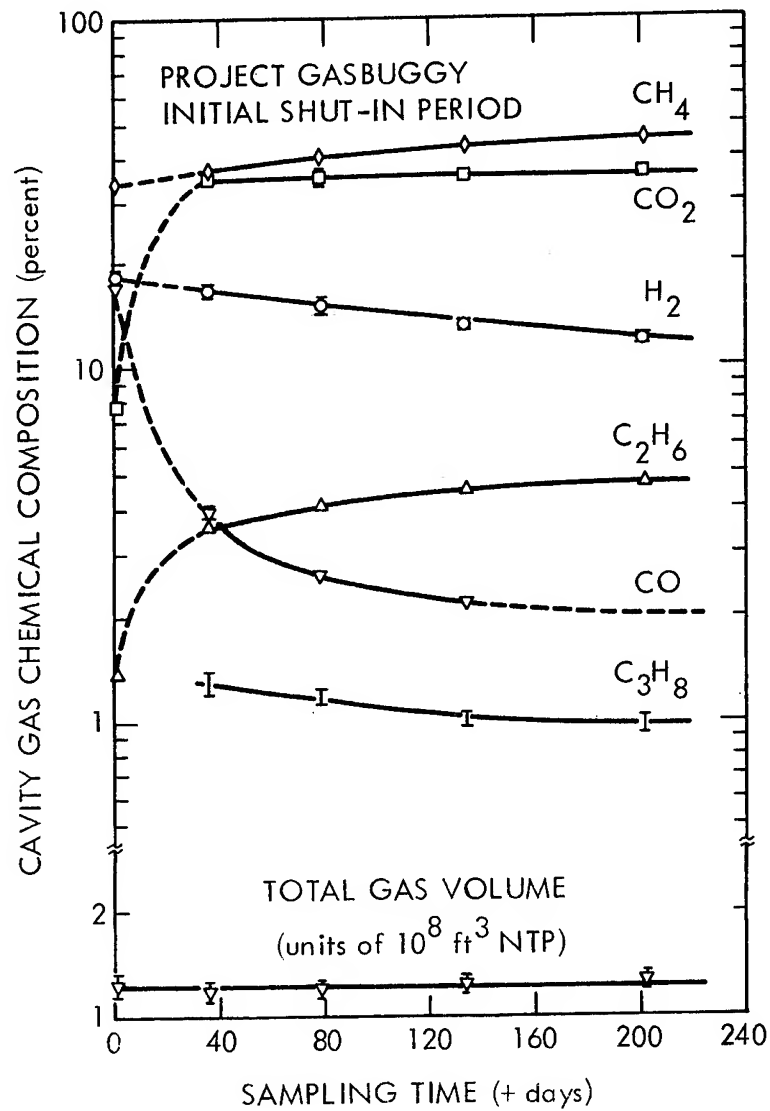


Fig. 6. Changes in the composition of the cavity gas as a function of time.

Table 1A. Chemical composition of Gasbuggy cavity gases.

Sampling time (+ days)	Mole percent				
	H ₂	CH ₄	C ₂ H ₆	C ₃ H ₈	CO
1	18.1 ± 0.6	34.0 ± 0.5	1.37 ± 0.02	—	17.1 ± 0.4
36	16.8 ± 0.4	36.9 ± 0.8	3.60 ± 0.06	1.3 ± 0.1	3.9 ± 0.2
79	15.2 ± 1.0	40.1 ± 2.0	4.1 ± 0.2	1.19 ± 0.04	2.6 ± 0.1
134	13.2 ± 0.1	43.2 ± 0.7	4.5 ± 0.1	1.02 ± 0.05	2.20 ± 0.04
203	12.0 ± 0.2	44.2 ± 0.7	4.7 ± 0.2	0.97 ± 0.05	—
					36.2 ± 0.7

Table 1B. Elemental concentrations in cavity gas.

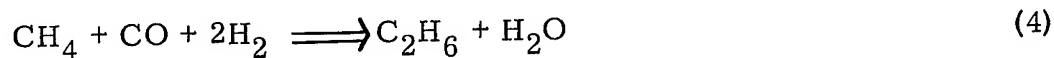
Sampling time (+ days)	Gram atoms/ft ³ NTP			Atom percent		
	H	C	O	Total	H	C
1	2.09	0.714	0.379	3.18	66.2	22.4
36	2.46	1.01	0.871	4.34	56.5	22.3
79	2.61	1.05	0.860	4.52	57.7	23.2
134	2.76	1.09	0.856	4.71	58.5	23.2
203	2.73	1.07	0.834	4.63	58.9	23.0
						18.1



which is observed to proceed slowly throughout the sampling period. In addition, natural gas from the formation has entered the cavity to maintain constant pressure. The water gas reaction (Eq. 1) is proceeding toward equilibrium. Adding the reactions represented by Eqs. 1 and 2 produces:



Note that CO_2 does not enter, and is indeed constant over the major sampling period. H_2 and CO are being used up, while methane is increasing. The observed increase in ethane concentration may be due to a reaction such as:



The observed decrease in propane may or may not be significant. Variation such as that seen can be attributed to fractionation of the sample during the later sampling periods.

Another way to view the chemical data is in terms of totals of elements in the gas. These are shown in Fig. 7. Plotted across the bottom of the figure is the total gas volume to provide a base line. Above it, in the center portion of the figure, are the concentrations of the atoms of interest. Because of the constancy of the total gas volume, these curves represent totals equally well. The curves appearing at the top of the figure are chemical composition in terms of atom percent.

Note that the total hydrogen atoms are actually increasing at early time. It is only because total atoms in the gas are increasing more rapidly that the decrease in hydrogen fraction is observed. This is just another way of saying that the chemical reactions are proceeding toward production of more complex molecules, and natural gas influx is being called on to maintain a constant volume of gas within the chimney.

Increase in oxygen at early times is due to the production of CO_2 by the water gas reaction (Eq. 1). Decrease at late times can be accounted for by invoking the reaction defined by Eq. 3.

The radiochemical results for tritium in hydrogen, methane, and ethane are plotted in Fig. 8 from the data summarized in Table II. Again total gas

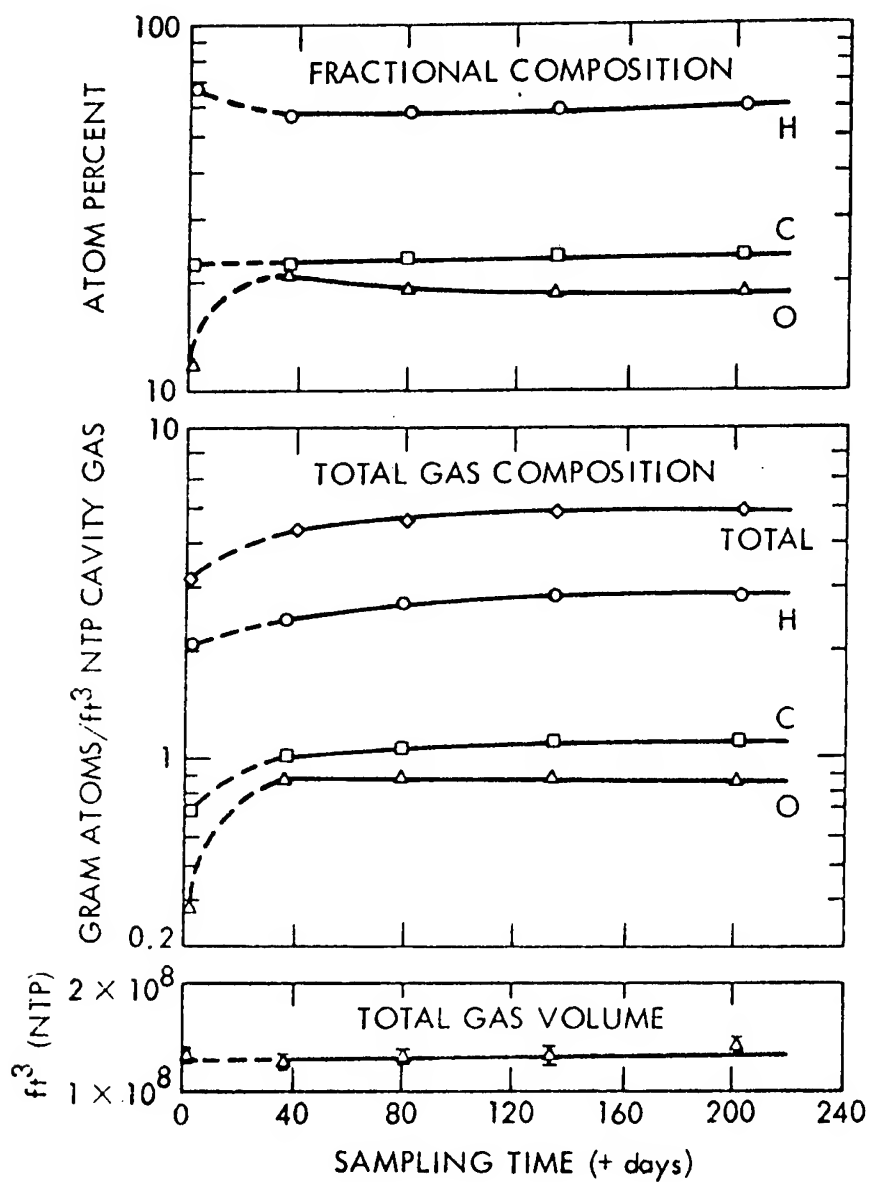


Fig. 7. Summary of elements present in the gas as a function of time.

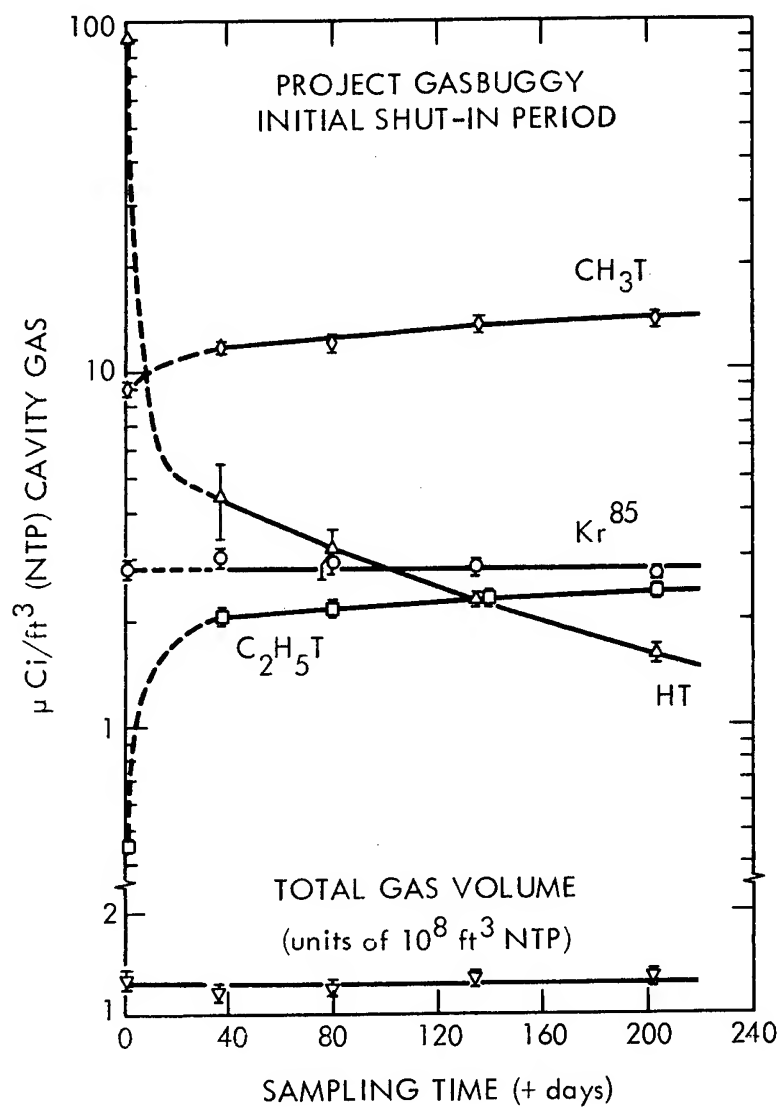


Fig. 8. Radionuclide concentrations plotted as functions of time.

Table II. Radionuclide concentrations of Gasbuggy cavity gases.

Sampling time (+ days)	Total gas volume 10 ⁸ ft ³ NTP	$\mu\text{Ci}/\text{ft}^3$ of cavity gas				Total tritium
		Kr ⁸⁵	HT	CH ₃ T	C ₂ H ₅ T	
1	1.25 ± 0.08	2.8 ± 0.2	91 ± 1	8.9 ± 0.1	0.45 ± 0.01	100 ± 1
36	1.17 ± 0.07	3.0 ± 0.2	4.4 ± 1.1	11.7 ± 0.1	2.06 ± 0.09	18 ± 1
79	1.20 ± 0.08	2.9 ± 0.2	3.1 ± 0.5	12.0 ± 0.7	2.15 ± 0.06	17 ± 1
134	1.24 ± 0.06	2.8 ± 0.1	2.3 ± 0.1	13.6 ± 0.7	2.3 ± 0.1	18 ± 1
203	1.28 ± 0.07	2.7 ± 0.1	1.6 ± 0.1	13.9 ± 0.7	2.4 ± 0.1	18 ± 1
Average	1.23 ± 0.04	2.8 ± 0.1				(18 ± 0.8)

volume is plotted along the base line for reference. This information was obtained quite simply by dividing the total Kr^{85} by the $\text{Kr}^{85}/\text{ft}^3$ NTP determined from radiochemical analysis of the samples. Total Kr^{85} is estimated at 350 curies according to the anticipated performance of the nuclear explosive. The observed gas volume of $1.2 \times 10^8 \text{ ft}^3$ NTP can be contained in $2.1 \pm 0.1 \times 10^6 \text{ ft}^3$ void at 150°F and 950 psig (the conditions observed on January 23, 1968). Such a void was estimated from data obtained during the production testing, implying that the total krypton-85 estimate is reasonable.

The actual Kr^{85} data seem to be trending toward lower concentrations, but because of analytical errors associated with the data no conclusion can be reached as to the significance of this trend. The best fit to the data within these uncertainties is probably the line shown.

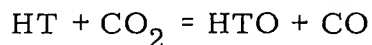
At early times a large fraction of the gaseous tritium existed in the form of hydrogen. The data point at 1 day corresponds to some 30% of the total of four grams of tritium assumed present in the postshot chimney. During the first month the HT level dropped quite rapidly and continued to decline at a slower rate. Prior to the time reentry of the chimney well was accomplished, tritiated methane became the principle contaminant of the chimney gas. About 5% of the total tritium remains gaseous at late times. Presumably the other 95% is in the form of water. No meaningful tritiated water results can be reported here. Obtaining a representative sample of water in the chimney gas is extremely difficult. The actual numbers range from about a microcurie per cubic foot of cavity gas to $1/1000$ of that value, the variation being due primarily to dilution of the water in the sample by non-tritiated water within the cavity and reentry well casing coming from the over lying aquifers.

I^{131} , a potential problem nuclide at early times, was not seen in any of the samples. An upper limit of 10^{-4} of this nuclide existing as a gas within the chimney appears conservative. No other radionuclides have been detected which would cause a problem at times longer than a few months. Ar^{37} produced by neutron activation of the calcium in the rock is the only other radionuclide now prominent in the gas. Its initial concentration was about $120 \mu\text{Ci}/\text{ft}^3$ NTP. Due to its half-life (35.4 d) the Ar^{37} concentration is now less than that of Kr^{85} .

Analysis of a sample obtained from GB-2R is not yet complete, but does indicate the presence of a small quantity of cavity gas in the formation out to at least 300 ft.

Changes in the concentrations of the tritiated species appear to follow the trends observed for chemical compositions, with the exception that the decrease of tritiated hydrogen is more marked than the corresponding decrease in hydrogen gas. Equilibrium reaction such as the water gas reaction (Eq. 1) provide a path whereby tritium can exchange with hydrogen in water, reducing the tritium concentration. The overall effect is that the ratio of tritium to hydrogen tends to equalize in all hydrogen-containing species participating in the exchange. The degree to which this is observed depends on the exchange rate.

The rapid initial decrease in HT concentration can therefore be explained using the water gas equilibrium:



and reactions such as Eqs. 2 and 4 can be used to explain the observed increases in tritiated methane and ethane.

Figure 9 compares specific activities for the tritiated species (data for this plot are tabulated in Table III). These curves demonstrate the trend toward a uniform tritium to hydrogen ratio. The HT/H₂ ratio is seen to drop quite rapidly as the water gas reaction approaches equilibrium and to change slope as the slower chemical reactions begin to dominate. The rapid decrease observed indicates that the T/H in cavity water was quite low, and is consistent with the production of CH₄ and C₂H₆ by reactions such as those defined by either Eq. 2 or Eq. 4.

The fact that both the CH₃T and C₂H₅T curves appear to be flat over the entire sampling period is not contradictory. They are being produced by hydrogen of nearly the same specific activity as they themselves have. In fact, the samples at 134 days show very nearly the proper 1:2:3 ratio for the three curves. Presumably the eventual downward trend should appear when the HT is further reduced by exchange with water. The fact that exchange equilibrium existed at ~134 d and that the HT/H₂ ratio is still decreasing may imply entry of non-tritiated water into the chimney.

Isotopic effects resulting from differences in bonding energies between tritium and hydrogen also tend to reduce tritium in hydrogen in favor of water at low temperatures.

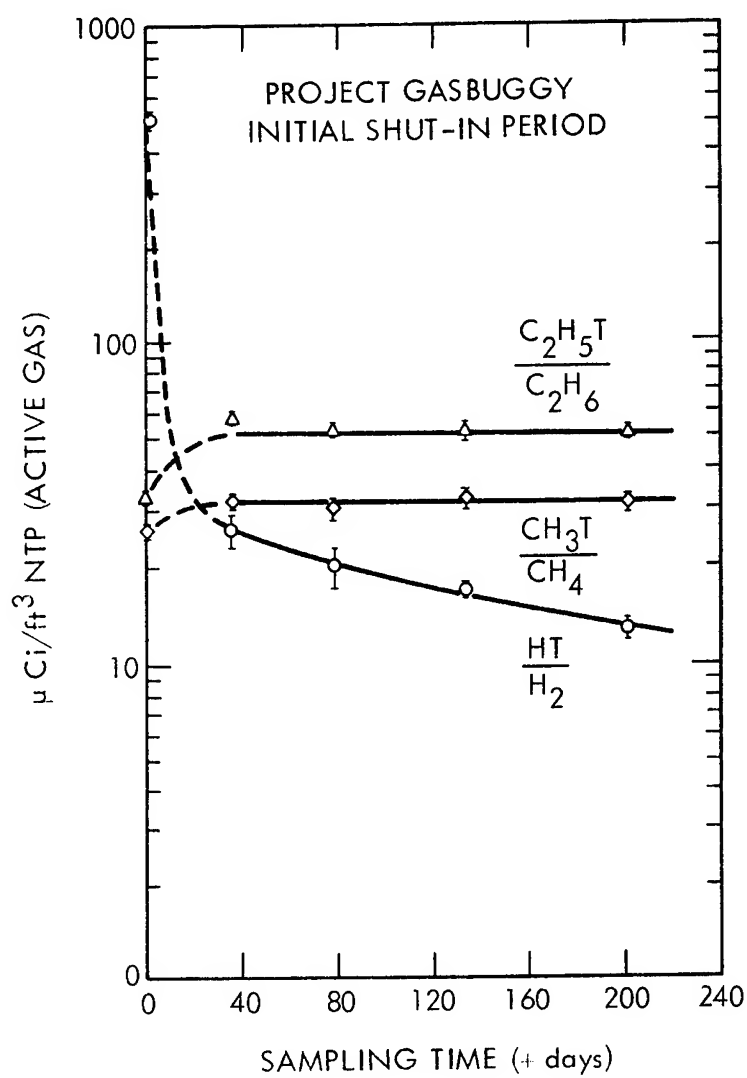


Fig. 9. Specific activities for the tritiated species as functions of time.

Changes in chemical composition of the gas during the 11-day flow test which occurred during June/July 1968 are plotted on Fig. 10 from the data tabulated in Table IV. Logarithmic increases with flow rate for components of natural gas are complimented by corresponding decreases for gases solely of cavity origin. Results plotted cover the flaring period at 5×10^6 ft³ per day nominal flow.

The composition of the produced gas is nearly like that of the cavity gas. About one third of the original cavity gas was removed by dumping two fifths of a cavity volume of gas. On the average only 17% of the produced gas came from outside the chimney. At this rate a factor of 10 reduction in

Table III. Specific activities of Gasbuggy cavity gases.

Sampling time (+ days)	$\mu\text{Ci}/\text{ft}^3$ NTP active gas		
	HT/H ₂	CH ₃ T/CH ₄	C ₂ H ₅ T/C ₂ H ₆
1	500 \pm 40	26.2 \pm 0.4	32.8 \pm 0.5
36	26 \pm 3	31.7 \pm 0.9	57 \pm 2
79	20 \pm 3	30 \pm 2	52 \pm 3
134	17 \pm 1	32 \pm 2	52 \pm 3
203	13 \pm 1	31 \pm 2	51 \pm 2

Table IV. Chemical composition of cavity gas produced from Gasbuggy. First drawdown test period was June-July 1968.

Sampling time (+ days)	Approximate total flow 10 ⁶ ft ³	H ₂	CH ₄	C ₂ H ₆	C ₃ H ₈	CO ₂
134	0	13.3	43.3	4.50	0.98	35.1
203	5	12.0	44.1	4.69	0.97	36.1
205	20	11.1	46.7	4.90	1.30	34.0
207	30	10.8	47.7	4.91	1.31	33.1
210	35	10.5	48.9	4.97	1.37	32.2
213	50	9.8	49.8	5.06	1.56	30.5

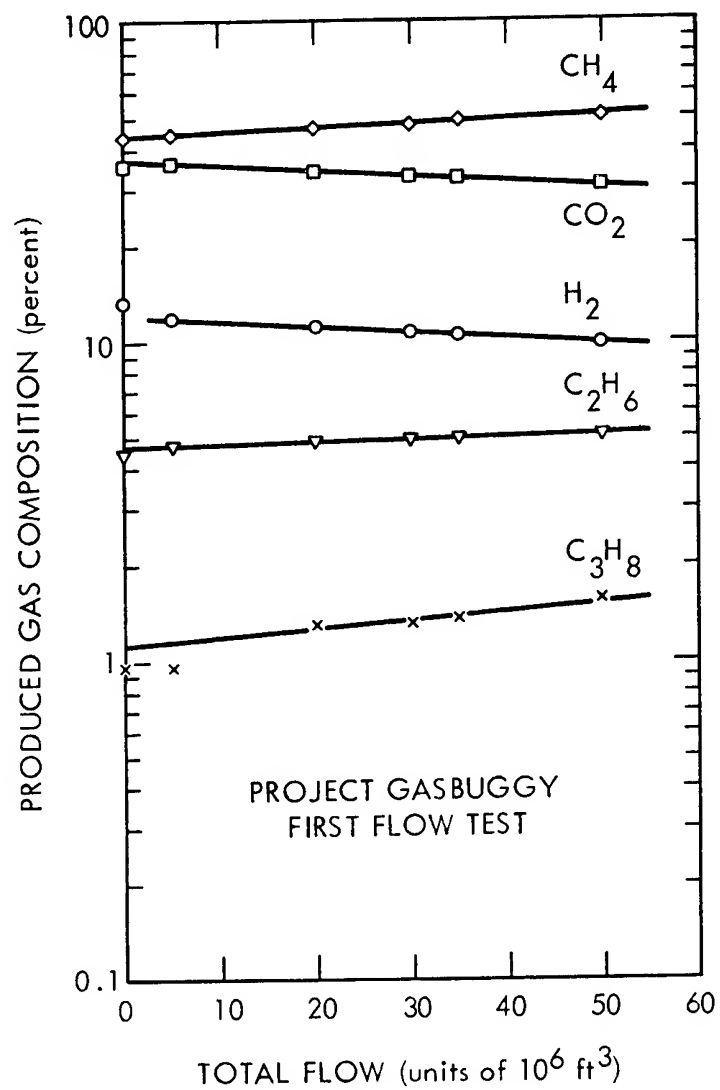


Fig. 10. Changes in chemical composition that occurred during the 11-day flow test.

contamination of the chimney gas can be achieved by flaring about two chimney volumes ($\sim 2.5 \times 10^8 \text{ ft}^3 \text{ NTP}$). Further experiments are needed to define the long term behavior of the cavity flushing, and to establish a consistent model for estimating the fraction of cavity gas removed as a function of flow rate.

Radiochemical analysis of these samples are not yet complete. Preliminary assessment of changes in concentration that have been observed do, however, seem to generally fit those shown here for CO_2 and H_2 . Four days of flaring at a nominal rate of $0.75 \times 10^6 \text{ ft}^3$ per day followed, but the analytical results were too late for inclusion in this report. They do, however, show a marked deviation from the lines established at the higher flow indicating much more dilution of the cavity gas by influx of formation gas. This observation corresponds to the observed increase in cavity pressure during the low flow rate flaring.

The gas quality program at LRL is continuing its investigations in an effort to gain a better understanding of the complex interactions of the Gasbuggy gas with itself and its environment. These results, and the interpretations which can be drawn from them, will be added to those presented here and made publicly available in the near future.

TECHNICAL AND SAFETY PROGRAM REPORTS
PROJECT GASBUGGY

A. TECHNICAL REPORTS - (already issued)

<u>Authoring Organization</u>	<u>Report No.</u>	<u>Report Title</u>
EPNG/AEC/USBM/LRL	PNE-1000	Project Gasbuggy (Feas. Study Rpt.)
LRL	PNE-1001	Pre-Shot Summary
LRL	PNE-1003	Preliminary Post-Shot Summary
EPNG	PNE-G-9	Drilling & Testing Operations
LRL	PNE-G-10	Gas Quality Investigation Program Status Rpt.
LRL	PNE-G-11	Post-Shot Geologic Investigation
USBM/EPNG	PNE-G-13	Status of Reservoir Evaluation

B. TECHNICAL REPORTS - (to be prepared)

SL	PNE-1002	Free-Field & Surface Ground Motions
LRL	--	Prediction & Results of Dynamic Effects
LRL	--	Analysis & Interpretation of Gaseous Radioactivities
LRL	--	The Gasbuggy Seismic Source
LRL	--	Response of the Navajo and El Vado Dams
EPNG/USBM/LRL	--	Reservoir Geology
EPNG/USEM	--	Post-Shot Flow Tests
EPNG/USBM	--	Reservoir Analysis

C. SAFETY REPORTS - (already issued)

NV	PNE-G-12	Operational Safety Aspects
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D. SAFETY REPORTS - (to be prepared)

EIC	PNE-1006	On-Site Radiological Safety
USPHS	PNE-1007	Off-Site Radiological Surveillance
ESSA/ARFRO	PNE-1008	Weather and Radiation Predictions
II	PNE-1009	Ground Water Safety Evaluation
ERC	PNE-1010	Analysis of Ground Motion & Containment
USBM (BuMines)	PNE-1011	Mine & Well Safety
JAB	PNE-1012	Structural Response
USGS	PNE-1013	Geology & Hydrology
USC&GS	PNE-1014	Seismic Measurements

E. ABBREVIATIONS OF ORGANIZATIONS

EIC	Eberline Instruments Corp., Santa Fe, N.M.
EPNG	El Paso Natural Gas Co., El Paso, Texas
ERC	Environmental Research Corp., Alexandria, Va.
ESSA/ARFRO	Environmental Science Services Administration/ Air Resources Field Research Office, Las Vegas, Nev.
II	Isotopes, Inc., Palo Alto, California
JAB	John A. Blume & Associates, San Francisco, Calif.
LRL	Lawrence Radiation Laboratory, Livermore, Calif.
NV	USAEC Nevada Operations Office, Las Vegas, Nevada
SL	Sandia Laboratory, Albuquerque, N.M.
USAEC	U. S. Atomic Energy Commission
USEM	Bureau of Mines, U. S. Department of the Interior
USC&GS	U. S. Coast & Geodetic Survey, Las Vegas, Nev.
USGS	U. S. Geological Survey, Denver, Colo.
USPHS	U. S. Public Health Service, Las Vegas, Nev.

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